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Characterisation, Quantity and Sorptive Properties of Microplastics Extracted From Cosmetics

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Abstract

Cosmetic products, such as facial scrubs, have been identified as potentially important primary sources of microplastics to the marine environment. This study characterises, quantifies and then investigates the sorptive properties of plastic microbeads that are used as exfoliants in cosmetics. Polyethylene microbeads were extracted from several products, and shown to have a wide size range (mean diameters between 164 to 327 μm). We estimated that between 4594 – 94500 microbeads could be released in a single use. To examine the potential for microbeads to accumulate and transport chemicals they were exposed to a binary mixture of ^{3}H-phenanthrene and ^{14}C-DDT in seawater. The potential for transport of sorbed chemicals by microbeads was broadly similar to that of polythene (PE) particles used in previous sorption studies. In conclusion, cosmetic exfoliants are a potentially important, yet preventable source of microplastic contamination in the marine environment.

Keywords:

- Microplastic; Exfolianting Microbeads; Polyethylene; Ocean pollution; Contaminant

1. Introduction

Plastics provide a diverse range of inexpensive, lightweight, strong, durable and corrosion-resistant products (Thompson *et al.,* 2009b). The success of plastics as materials has been substantial and they are used in a wide range of applications. This versatility, together with their low cost, has resulted in the annual worldwide production of around 300 million tonnes (Plastics Europe. 2014). Approximately 50% of production is used to make packaging, much of which is used in disposable applications. This creates a major waste management problem, with plastics accounting for approximately 8-10% of all the waste generated in the UK (Barnes *et* al., 2009; Hopewell *et al*., 2009).

Around 700 species of marine organism have been reported to encounter marine debris in the natural environment, with plastic debris accounting for over 90% of these encounters (Gall and Thompson, 2015). Large plastic items, such as discarded fishing rope and nets, can cause entanglement of invertebrates, birds, mammals, and turtles (Carr, 1987; Eerkes-Medrano et al., 2015; Fowler, 1987; Laist, 1997) but the marine environment is also contaminated with much smaller microplastics particles (defined by NOAA as <5mm). These have been reported at the sea surface (Law and Thompson 2014), on shorelines (Claessens et al., 2011), and on the sea bed (Van Cauwenberghe et al., 2013). The sources of microplastics

include fragmentation of larger items (secondary sources), and direct inputs of microplastic sized particles, such as microbeads used in cosmetics and pre-production pellets (primary sources). It is important to understand the relative importance of these sources as well as the size and abundance of microplastic particles released, since this will influence encounter rate and availability to biota (Cole et al 2011; Teuten et al., 2007; Thompson et al., 2009a).

There is growing evidence that the amount of microplastics in marine waters is increasing, with unknown ecotoxicological consequences (Goldstein et al., 2012). Fendall and Sewell (2009) reported on microbeads used as “scrubbers” in cosmetics products, which they described as being up to 500 μm in diameter, being released into the natural environment and potentially available to organisms. Ingestion of microplastics, has been reported for a wide range of marine organisms including deposit and suspension feeders (Browne et al., 2008; Graham and Thompson, 2009), crustaceans (Murray and Cowie, 2011), fish (Boerger et al., 2010), marine mammals (Denuncio et al., 2011), and seabirds (Avery-Gomm et al., 2012; Van Franeker et al., 2011). However, the extent, if any, to which chemicals sorbed onto, or incorporated into plastics can desorb from plastic particles, and transfer to the tissues of marine organisms is less clear. Recent experimental trials provide evidence for the role of plastics in the transfer of chemicals with subsequent adverse physiological effects (Besseling et al., 2013; Rochman et al., 2013), but studies based on bioaccumulation models concluded that the transfer of contaminants from plastics to marine organisms upon ingestion is of limited importance compared to other pathways (Gouin et al., 2011; Koelmans et al., 2013).

Microplastics have been used to replace natural exfoliating materials (for example, pumice, oatmeal, apricot or walnut husks) in cosmetics and have been reported in a variety of products such as hand-cleansers, soaps, toothpaste, shaving foam, bubble bath, sunscreen, shampoo and facial scrubs (Fendall and Sewell, 2009; Gregory, 1996; Zitko and Hanlon, 1991; UNEP, 2015).

Industry uses the terms ‘microbeads’ to describe microplastic particles present as ingredients in personal care and cosmetic products; they may also be called microspheres, nanospheres, plastic particulates (UNEP, 2015). Around 93% of the ‘microbeads’ used in cosmetics are polyethylene (PE), but they can also be made of polypropylene (PP), PE terephthalate (PET), polymethyl methacrylate (PMMA) and nylon (Gouin et al., 2015; Eriksen et al., 2013; UNEP, 2015). Microbeads are likely to be transported to wastewater treatment plants, where some will be captured in oxidation ponds or sewage sludge. However, due to their small size, it is anticipated that a substantial proportion will pass through filtration systems and enter aquatic environments (Fendall and Sewell, 2009).

Leslie et al. (2013), examined wastewater treatment plants that discharge into the North Sea, the Oude Maas River or the North Sea Canal and reported that the treated effluent contained on average 52 pieces of microplastics/L. Eriksen et al.

115 (2013) also reported substantial amounts of multi-coloured microplastic spheres in
116 surface waters of the Laurentian Great Lakes of the United States which were
117 suspected to originate from consumer products. This provides evidence that
118 microplastics are not all captured in sewage sludge of wastewater treatment plants
119 and is of broad concern, since treated effluent from sewage disposal sites is
120 discharged into a range of water bodies, including into inland waters, estuaries and
121 the sea (DEFRA, 2002).

122

123 Gouin et al. (2011) estimated that the per capita consumption of microplastic used in
124 personal care products for the U.S. population, based on the usage of PE
125 microplastic beads used in personal care products, was approximately 2.4 mg per
126 person^{-1} per d^{-1}, indicating that the U.S. population may be emitting an estimated 263
127 tonnes per yr^{-1} of PE microplastic (Gouin et al., 2011). To set this into perspective, in
128 terms of its contribution to marine litter, this annual quantity is approximately
129 equivalent to 25% of the total mass of plastic that is estimated to have accumulated
130 in the North Atlantic Subtropical Gyre (Law et al., 2010; Gouin et al., 2011).

131

132 Facial scrubs are one type of cosmetic which contains microplastics as exfoliating
133 agents. Due to this, such products could contribute microplastics contamination to
134 the marine environment. Despite concerns about the potential for products
135 containing microbeads to represent a major source of microplastics to the
136 environment, only one study has measured microplastics in facial scrubs (Fendall
137 and Sewell, 2009), and there are no peer reviewed publications confirming the type
138 or quantity of microplastic polymers used in facial scrubs. Here we examined six

brands of facial scrubs manufactured by three companies and describe the microplastics (plastic microbeads) present, in terms of polymer type, colour, size, weight and abundance. We also investigated the sorptive properties of the microplastics in relation to the potential for transport of the POPs phenanthrene (Phe) and dichlorodiphenyltrichloroethane (DDT) and compared them with commercially available PE particles previously used in adsorption/desorption studies of persistent organic pollutants (POPs) (Bakir et al., 2012, 2014a, b; Teuten et al., 2007).

2. Methods

2.1 Sample preparation

Six major brands of facial scrubs were chosen, based on their prevalence in major supermarkets close to Plymouth UK. All of the products listed in their ingredients that they contained PE. Four replicates of each product were purchased, with each replicate sourced from a different supermarket to provide a representative sample.

Since the specific brand names of the products are not of particular relevance, they were labelled A-F.

Each facial scrub was a viscous liquid (A to D contained 150mL of product, E contained 125mL). The contents were subjected to vacuum filtration to obtain the plastic particles. The procedure required mixing each product in approximately 1L of boiling water, followed by vacuum filtration over Whatman Nº4 filter paper, then drying at 30°C to constant weight. Once dry, the particles were weighed by Precisa

2200C weighing scales and the residues were transferred into separate glass vials. A Kruskal-Wallis test was performed on the data, using R studio, to test whether the amount of microplastics per unit volume extracted differed between products ($p < 0.05$). This was followed by a *post-hoc* Nemenyi-Test to find which specific products significantly differed.

2.2 Visualisation and identification

Microplastics from each product were identified using Fourier transform infra-red spectroscopy (FTIR), using a Hyperion 1000 microscope (Bruker) coupled to an IFS 66 spectrometer (Bruker). The spectra obtained were compared to a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATRlibrary).

Some non-plastic residues were extracted and separated from the plastic particles using Endecotts woven wire sieves of varying mesh size. The mass of plastic particles was recorded.

A Malvern Mastersizer 2000 laser particle sizer (MM2) was used to measure the size-frequency distributions (SFDs) of the extracted plastic into sixty-eight different sized bands with logarithmic spacing (range 0.015 µm, to 2000 µm; Woolfe and Michibayashi, 1995). The resultant particle size distributions were expressed as a

183 volume weighted mean from an average of twenty five measurements per product.
184 The mean for each product was then calculated.

185

186 The number of plastic particles in each product, *N*, was estimated, assuming the
187 particles were of spherical shape, using the following equations:

188 i) $Vt = \frac{Mt}{D}$ ii) $V(avg\ particle) = \frac{4}{3}\pi r^3$ iii) $N = \frac{Vt}{V(avg\ particle)}$

189 where *Vt* is the total volume of plastic extracted, *Mt* is the total mass of plastic
190 extracted, *D* is the density, *V(avg.p)* is the mean volume of one particle, *N* is number
191 of particles, and r is the radius.

192

193 For each product: equation i) allowed calculation of the total volume of microplastic
194 extracted; equation ii) allowed calculation of the average volume of a microplastic
195 particle from each product; by dividing the total volume of microplastic by the
196 average volume of a microplastic particle, equation iii) allowed calculation of the
197 approximate number of particles in each product. Particles were then visualised by
198 scanning electron microscopy (JEOL, 7001F), imaging to describe both whole
199 particles and their topography.

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207 2.3 Sorption of pollutants to plastics

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209 As part of a separate, but related study, microbead exfoliants were extracted from
210 shower gel and used to examine the adsorption of POPs by microbeads. The
211 microbeads from the shower gel products were extracted and identified by FTIR
212 following the same methods in sections 2.1 and 2.2. As these microbeads were
213 extracted from different brands of exfoliant products, they are labelled X, Y & Z.
214 These microbeads were exposed to Phe and DTT; the results were then compared
215 with sorption to ultra-high-molecular-weight (UHMW) PE particles used in a previous
216 sorption study (Bakir et al., 2014a, 2014b, 2012).

217

218 Adsorption experiments were conducted in an ISO9001 accredited radioisotope
219 facility at the Plymouth University. ^{3}H-Phe and ^{14}C-DDT were selected as
220 contaminants in this study to allow simultaneous quantification and to compare with
221 past studies (Bakir et al., 2012). 10 mg of either UHMW PE or the extracted
222 microbeads were placed into three glass centrifuge tubes (50 mL) and 5 μL of ^{14}C-
223 DDT and 16 μL of ^{3}H-Phe were added to the walls of the tubes. The solvent was
224 allowed to evaporate and 25 mL of seawater (35 psu, 59.3 ± 0.26 mS) was added
225 and the tubes were equilibrated for 48 hours (Bakir et al., 2014a) in the dark at 18 ºC
226 under continuous horizontal, rotary agitation at 220 rpm. All experiments were
227 carried out in triplicate. The concentration of contaminant was determined in the

aqueous and solid phase by counting the β decay from the ^{14}C-contaminant by liquid scintillation counting (LSC) as outlined in Bakir et al. (2012). The amount of contaminant in each phase was quantified using a calibration curve prepared by counting known amounts of the contaminant.

The single point distribution coefficient, single point K_d, was calculated using the equation:

(iv) $$Kd = [q_e]_{solid} / [C_e]_{aq.}$$

where q_e is the amount of contaminant adsorbed onto plastic (μg kg^{-1}) at equilibrium and C_e is the contaminant concentration in the aqueous phase at equilibrium (μg L^{-1}).

2.4 Statistical analysis

A two-factor ANOVA, with contaminants and the microbead type considered as fixed factors, was used to characterise any significant differences ($p < 0.05$) between the distribution coefficients calculated from the sorption of Phe and DDT onto microbeads. Cochran's test was used to ensure that the data fulfilled the pre-requisites for parametric analysis and the appropriate data were ln(x+1) transformed. Student-Newman-Keuls (SNK) tests were then used to identify any significant terms. The tests were carried out using GMAV5 software (Underwood et al., 2002) and are presented in the supplementary information.

3.0 Results

3.1 Extraction and Identification

All of the products contained microplastic particles of PE, which was in agreement with their stated ingredients. Product C also contained green and yellow particles that were slightly larger than the PE microbeads. These could not be identified by FTIR using the Bruker spectral database and were removed from the samples via sieving and are not included in any of the calculations. The collected solids from product C also contained micro-'glitter'. These 'glitter' particles were small and could not be removed from the filter paper for further analysis. However, 'glitter' is commonly manufactured from plastic, such as PE.

The weight of microplastic extracted varied significantly between products (Kruskal-Wallis test, $p = 0.0012$, Fig 1); the products which were significantly different from each other were C and E ($p = 0.0009$); D and E ($p = 0.0463$) (*post hoc* Nemenyi-Test).

3.2 Size-Frequency Distributions

Microplastics from the facial scrubs showed polydispersed size ranges, each with logarithmic bimodal distributions (Fig 2). Product B had the largest size range (10μm to >2000 μm), whereas product A was the most homogenous, ranging from 8μm to 56μm, with the largest proportion of smaller particles. Size frequency by volume distributions were used to calculate the mean diameters for each product. Products D-F had similar volume-weighted mean diameters, which were 288.80 μm, 289.63 μm and 293.48 μm respectively. The particles in product B and C were larger, with mean diameters of 326.83 μm and 317.91 μm, while product A was much smaller with a mean diameter of 163.82 μm. The volume-weighted mean diameters were used to estimate the number of particles in each product. Since the absolute density of the extracted plastics was not known, we calculated estimates using a range of standard densities. For PE these were, high (0.959 g/cm^3), medium (0.940 g/cm^3) and low density (0.910 g/cm^3).

Particle diameter, rather than the average weight in each product, was found to have the greatest effect on abundance estimates. Product E had on average 11.47 g of PE in each bottle, with a mean particle size of 289.63 μm, resulting in an estimated 6423 particles per mL. Whereas product A had less PE by weight with, on average,

6.11g in each bottle, but resulted in an estimate of 18906 particles per mL because the mean size was smaller (163.82 μm); being the highest quantity in any of the products. Product C had the second largest PE particles (317.91 μm), but the lowest particle abundance, with only 919 particles per mL. This data implies that the products tested could each contain between 137,000 and 2,800,000 microparticles (Figure 3). The quantity of particles was calculated using data for the volume mean diameter, however the size particle distribution had a tail of smaller particles, hence the particle abundances calculated are likely to be underestimates.

The shape and surface topography of the extracted microplastic particles was visualised by scanning electron microscopy. For all the brands, the extracted microplastics had a variety of shapes, including ellipses, ribbons, and threads, as well as irregular fragments (Fig 4). An exception was product F, which in addition to irregular shaped pieces, also contained smooth, blue, PE spheres that were substantially larger than the rest of the particles, but represented a small proportion of the total amount of plastics present. Some of these spheres were fragmenting (Fig 4).

The colour of microplastics used in the different products also varied (Table 1). All products contained white microplastics, but products A, D, E and F also contained coloured particles. The coloured microplastics in products D-F were larger than the white plastics, but were less abundant. The white and pink microplastics in product A were of similar size to each other.

317 3.3 Sorption of persistent organic pollutants

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319 Visualisation of microbeads extracted from products X, Y, and Z showed they could
320 be differentiated between “smooth” and “rough” forms. This particle shape
321 differentiation was also observed in products A-F, where A-E contained "smooth"
322 particles and product F contained both "smooth" and "rough" forms (Fig 4). Therefore,
323 we considered sorption onto both morphologies. Results showed that microbeads
324 extracted from the cosmetic products were able to sorb Phe and DDT from seawater
325 (Fig 5). Sorption capacity for all plastics was significantly higher for DDT compared
326 to Phe ($p < 0.05$, Table 2). The “rough” microbeads were more efficient at adsorbing
327 POPs from seawater than “smooth” ones, probably due to increased surface area.
328 The “rough” microbeads were also more similar in shape, surface texture and
329 sorptive property for POPs to PE particles used in previous experiments (e.g. Bakir
330 et al., 2012, 2014a, b; Teuten et al., 2007). There were some significant differences
331 between adsorption by microbeads and adsorption by PE particles and the direction
332 of these effects was that microbeads from cosmetics tended to adsorb lower
333 concentrations of POPs then PE particles. However, broadly speaking, it would
334 appear that results from previous studies on transport of chemicals by sorption on to
335 plastic are comparable with the transport potential on microbeads.

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4.0 Discussion

Microplastics found within cosmetics such as facial scrubs, will routinely be washed into sewers as a direct consequence of consumer use. Due to their size, a considerable proportion is likely to pass through preliminary sewage treatment screens (typically coarse, >6 mm, and fine screens, 1.5–6 mm) (Water Environment Federation, 2003). Effulent containing the microplastics would then be discharged into inland waters, estuaries and the oceans. A recent study reported that treated effluent from three sample sites in the Netherlands contained on average 52 microplastic particles/L (Leslie et al., 2013).Microbeads used as exfoliants in facial scrubs are likely to be an important primary source of microplastics contamination, due to the quantity of plastic used in each product.

When considering the potential consequences of the release of microbeads to the environment, if any, it is important to consider both the mass of plastic, and the number and size of the particles; the latter will have direct influence on the probability of encounters with wildlife.

The common application of facial scrub exfoliants is once per day, and it has been estimated that they are used by around 1.1 million women in the UK (Statista, 2013). Focussing on the products used in this study (A-F), and assuming that the typical daily amount used is 5mL, between 4594 – 94500 microplastic particles would have the potential to pass into the sewage system per use.

362 In terms of the mass of plastic entering the marine environment, previous work by
363 Gouin et al (2011) estimated that users in the U.S emit 2.4 mg of PE person $^{-1}$.d $^{-1}$,
364 amounting to an emission of 263 tonnes yr^{-1}. This estimate is calculated from data
365 on liquid soap consumption, and assumes that only 15% of the market is shared by
366 companies that use microplastic beads in their liquid soaps. However, many brands
367 do use exfoliating microbeads. Assuming that three out of four body exfoliants
368 contain microplastics (Marine Conservation Society, 2012), and that an estimate that
369 25% of the microplastic is caught by the sewage system, the UK population could
370 emit to the natural environment 40.5 – 215 mg of PE person $^{-1}$.d $^{-1}$, or between 16
371 and 86 tonnes yr^{-1} (population of the UK in 2013: 64.1 million, (The World Bank,
372 2013) just from facial exfoliants. In order to set these quantities into context, by way
373 of comparison, between 2009 and 2014 inclusive, in its annual weekend beach clean,
374 MCS typically collect around 9 tonnes of litter per year (over an average length of
375 115km of UK shoreline).

376 The presence of microplastics in sewage sludge has been reported previously by
377 Browne et al. (2011), who found that former sewage disposal-sites on the seabed in
378 UK waters contained more microplastics than non-disposal reference sites,
379 highlighting the potential for microplastics to accumulate in aquatic habitats. The
380 occurrence of microplastics within the marine environment is now well documented
381 in the water column, at the sea surface and sediments (Law and Thompson 2014).
382 Microplastics also account for around 10% of all reports of ingestion of marine
383 debris, highlighting their importance as a component of marine debris (Gall and
384 Thompson, 2015). Their size makes them accessible to organisms with a range of
385 feeding methods, including: filter feeders (mussels, barnacles), deposit feeders
386 (lugworms) and detritivores (amphipods, sea cucumbers) and zooplankton (Wright et

al., 2013a; Graham and Thompson, 2009; Thompson et al., 2009; Browne et al., 2008). However, studies that quantify the abundance of microplastic predominately report elongated fibres. This may in part be due to the relative ease of detection of pieces with these shapes, since they differ from many natural particles found in sediments. Hence, the prevalence of microplastics with non-fibrous shapes (Fig 4), for example microbeads from facial scrubs, may be under-reported in environmental sampling (Desforges et al., 2014; Lusher et al., 2014; Gallagher et al., 2015).

There is no way of effectively removing microplastic contamination once it is in the environment. The materials are too dispersed, the scale is too vast, ecological damage would be caused by any remediation (tiny organisms would likely be removed along with the microplastics), and the costs would be extremely high (UNEP, 2015). Since plastic is highly resistant to degradation, the abundance of microplastics in the ocean is assumed to be increasing, thus increasing the probability of ingestion by biota (Law and Thompson, 2014). The majority of microplastics extracted from the facial products herein were white or blue. It has been suggested by Wright, et al. (2013b) that these colours are similar to various types of plankton, a primary food source for surface feeding fish, which are visual predators.

A further potential problem associated with microplastics contamination is the possibility of transport of hydrophobic contaminants by microplastics: such contaminants have been found to sorb onto their surface of plastics and may transfer to biota upon ingestion (Avio et al., 2015; Bakir et al., 2014b; Teuten et al., 2007).

411 Previous studies have shown that PE particles have the potential to sorb and
412 concentrate a range of hydrophobic contaminants. This is of interest because these
413 contaminants can be released in conditions resembling those in the gut of an
414 organism (Bakir et al., 2014b) . However, at present, the environmental importance
415 of plastics as a vector in the transport of contaminants is not known. Here we show
416 that microbeads were able to adsorb greater amounts of DDT than Phe when both
417 chemicals were present in a mixture. This was in agreement with previous work
418 indicating that plastic showed a preferential affinity for DDT when present with Phe in
419 a binary mixture (Bakir et al., 2012). The size and shape of microbeads was also
420 found to be an important factor in their sorptive property for POPs and smooth
421 microbeads were found to adsorb lower concentrations of POPs than rough ones.
422 Rough microbeads were found to be most similar in their sorptive properties for
423 POPs to commercially available PE used in chemical transport studies (e.g. Bakir, et
424 al., 2012, 2014b; a; Teuten, et al., 2007). However, both types of microbeads were
425 broadly similar in their sportive properties to the microplastics used in previous
426 studies. Hence, on the basis of the experimental work here, it seems likely that
427 conclusions regarding the potential role of microplastics as possible vectors in the
428 transport of POPs in the environment could also be applied to transport by
429 microbeads from cosmetics.

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431

432 Rochman et al. (2013) investigated the transfer of hydrophobic organic compounds
433 (PAHs, PCBs and PBDEs) from PE to the fish, Japanese medaka (*Oryzias latipes*)
434 and the subsequent health effects. Plastic particles were exposed to natural marine

conditions, as opposed to laboratory exposures used in most previous studies. Environmental exposure will be highly dependent on the sites selected, which can be prone to variation. Results suggested the ingestion of virgin PE particles caused physiological stresses. However, the ingestion of contaminated PE particles led to the transfer of adsorbed contaminants, causing liver toxicity and pathology (Rochman et al., 2013). Laboratory studies using microplastic particles of polystyrene (Besseling et al., 2013) and PVC (Browne et al., 2013) have also indicated the potential for transfer of harmful chemicals with subsequent effects on biota. The present study showed that plastic particles present in cosmetics can be of varying size and shape and have differential affinities for sorption of POPs. Further work would be needed investigate the presence of chemicals such as pigments and dyes in microbeads, and their potential, if any, for migration from the polymer in either water or gut conditions.

The uneven topography of microplastics used in cosmetics could also provide habitats for diverse communities of microorganisms. A study by Zettler et al. (2013) described the presence of a rich eukaryotic and bacterial microbiota living on PE microplastic samples collected from the North Atlantic subtropical Gyre. Scanning electron microscope (SEM) images showed microbial cells embedded in pits on the plastic surface, and suggested that some members of this community could be accelerating the physical degradation of plastic; however this remains to be confirmed. The communities found on the plastic particles were distinct from surrounding surface water, indicating that plastic provides a novel habitat. Other studies have highlighted the potential for microplastic to act as vectors for microbial pathogens (Harrison et al., 2014).

460 Currently, there are reported to be eighty facial scrubs in the UK market, which
461 according to their product labelling, contain plastic material amongst their ingredients
462 (Beat the Microbead, 2015). However, some companies have indicated that they will
463 voluntarily phase out microplastics from their products. This could possibly be due to
464 research indicating the negative consequences of microplastics within the
465 environment; Fendall and Sewell (2009) stated that the presence of microplastics in
466 facial cleansers, and their potential use by millions of consumers world-wide, should
467 be of increasing concern, whilst Andrady (2011) also reported that there is an urgent
468 need to assess the future impact of increasing microplastics levels on the world's
469 oceans. There have also been associated public awareness campaigns (eg. Beat
470 the Microbead and Scrub it Out), urging consumers to boycott such products.
471
472 However, for the global market, usage statements vary within and between
473 companies, with some stating they will remove all microplastics from all their
474 products, while others say only PE will be removed. In some regions, legislation has
475 been introduced; for example, Illinois and California (U.S.A) have banned the
476 manufacture and sale of cosmetics that contain plastic microbeads, with similar
477 legislation being proposed for New York, Michigan, and Ohio (but not yet adopted)
478 (Driedger et al., 2015).
479
480 In conclusion, the present work characterised the microplastics in facial scrubs by
481 describing the polymer type, colour, size, weight and abundance. This allowed for
482 estimation that between 4594 and 94500 particles could be released into the
483 environment per use. We also estimate that the UK population is emitting 40.5 – 215
484 mg of PE person $^{-1}$.d $^{-1}$, resulting in a total of 16-86 tonnes yr^{-1}. Particle size, rather

485 than the average weight in each product, was found to be important as it had the
486 greatest effect on abundance estimates. Their small size also renders microbeads
487 accessible to a wide range of organisms, and may facilitate the transfer of
488 waterborne contaminants or pathogens. There are alternatives to the use of plastics
489 as exfoliating particles (UNEP 2015); hence these emissions of microplastic are
490 avoidable. Given the quantities of plastic particles reported here, and current
491 concerns about the accumulation of microplastics in the ocean, it is important to
492 monitor the extent to which manufactuers do voluntarily opt to remove microplastics
493 from their products. Such monitoring will help to establish whether there is a need for
494 further legislation.

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Tables

Product	Colour of Microplastic Present
A	White and Pink
B	White
C	White
D	White and Light Blue
E	White and Dark Blue
F	White and Dark Blue

Table 1. *Colour of microplastics found within six facial scrub products.*

Particle type	POP	Aqueous phase	Glass wall	Solid phase	Total recovery
Product X beads	DDT	12	8	59	78
	Phe	43	1	24	68
Product Y particles	DDT	7	8	91	106
	Phe	13	3	65	81
Product Z beads	DDT	20	26	33	79
	Phe	64	2	6	73
Product Z particles	DDT	3	8	90	101
	Phe	11	5	60	75
UHMW PE	DDT	2	6	87	94
	Phe	7	2	80	89

Table 2. *Recovery (%) of phenanthrene (Phe) and DDT following sorption experiments onto PVC and PE (average values displayed, n = 3).*

Figures

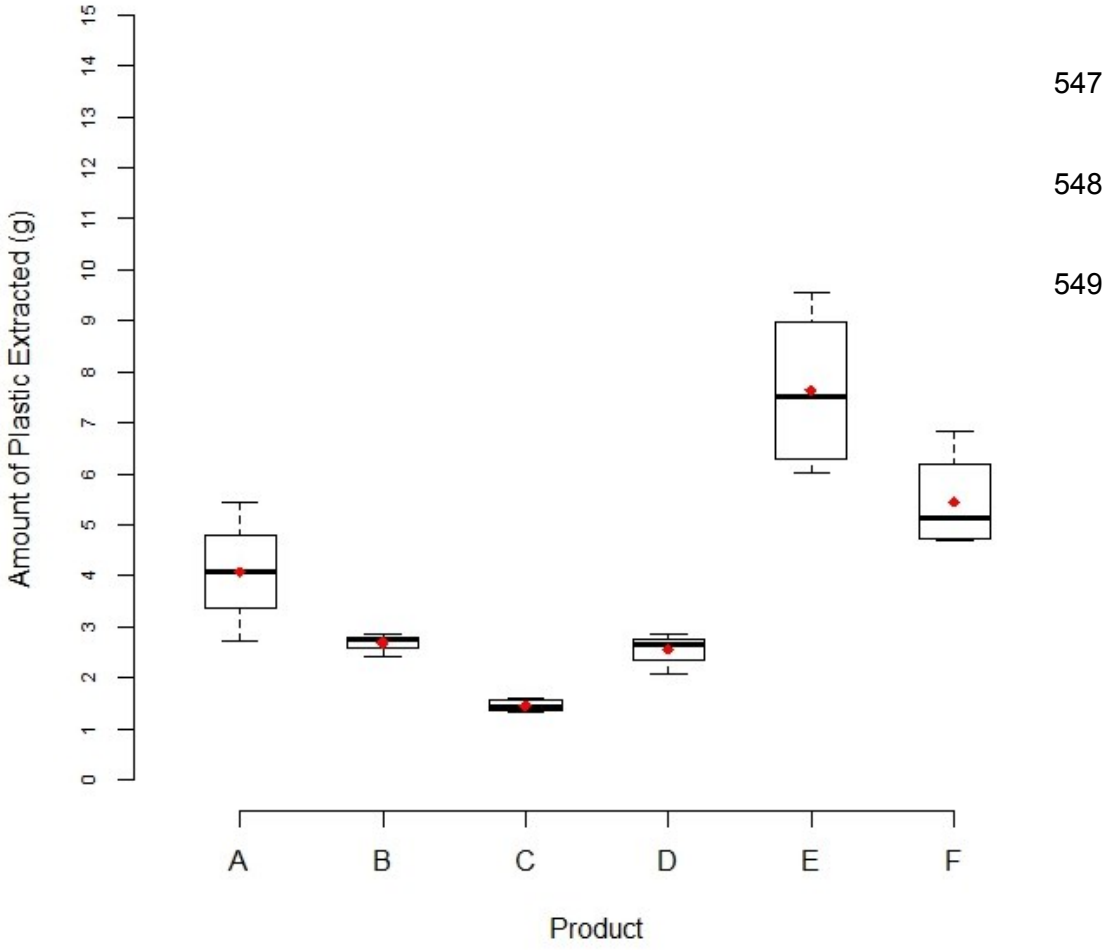
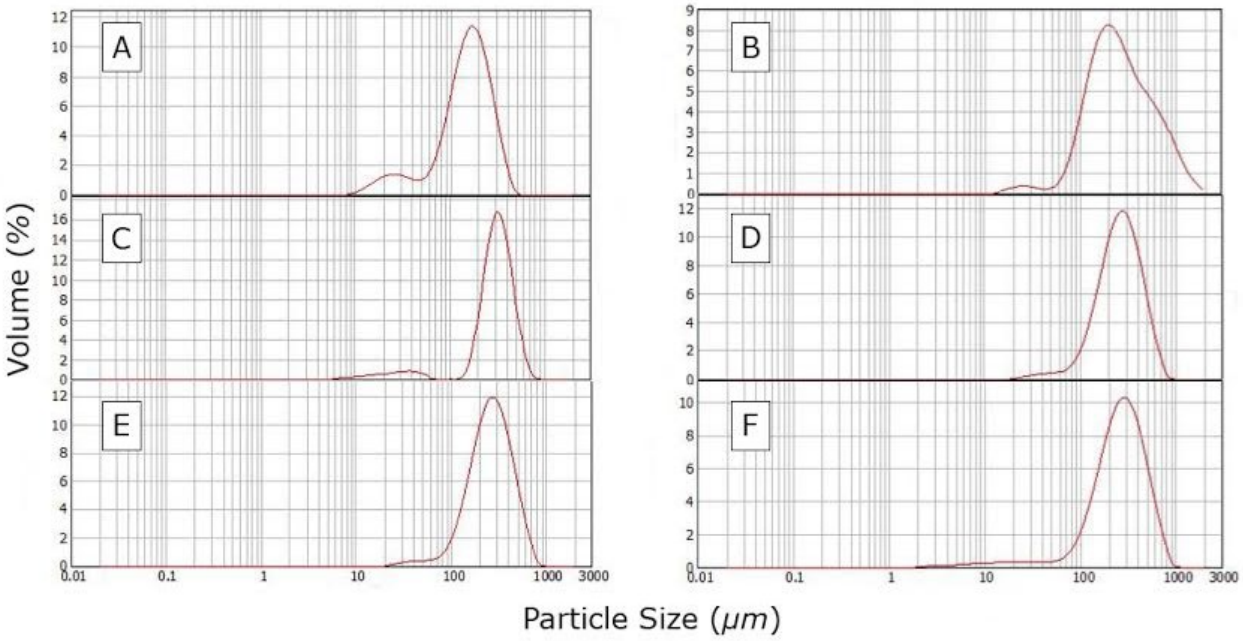


Fig 1. *Total mass of plastic microbeads extracted from six facial scrubs (A-F) per 100 mL.* Diamond symbol indicates $\bar{x}$ (n=4). The tails show both the maximum and minimum mass obtained, and the box represents the upper and lower quartiles. There were significant differences between the amount of microplastic in each of the products ($p < 0.05$).



555

556 **Fig 2.** *Particle size distribution of PE microbead particles extracted from six facial*
557 *scrubs (A-F).* Determined using a Malvern Mastersizer 2000, laser particle
558 sizer.

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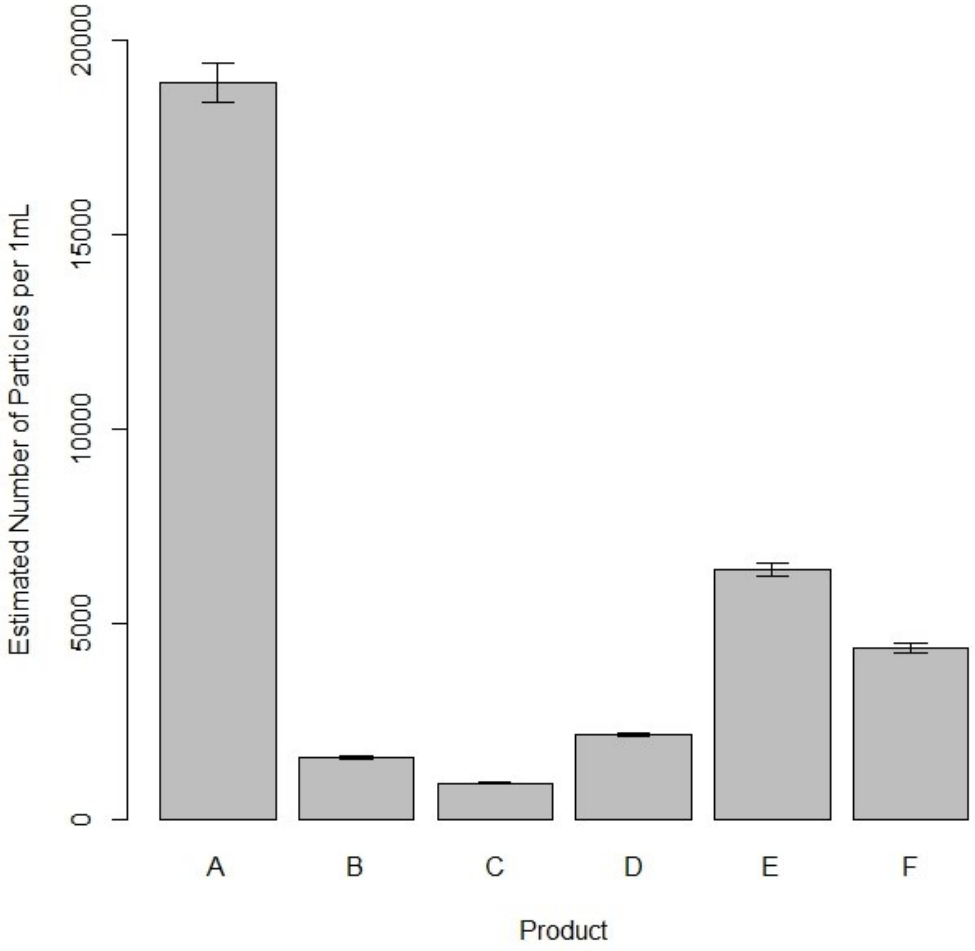
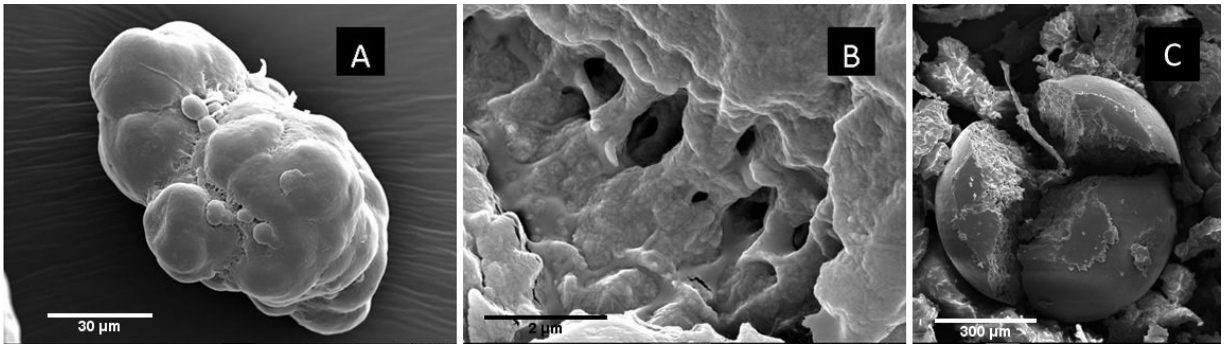


560

561 **Fig 3.** *Estimates for the number of PE microbead particles in six brands of facial*
562 *scrubs per 1mL*. Calculated using data from the volume weighted mean (n =
563 3, ±SD; correlating to the spread of the different amounts of particles
564 calculated for high, medium and low density PE).

565

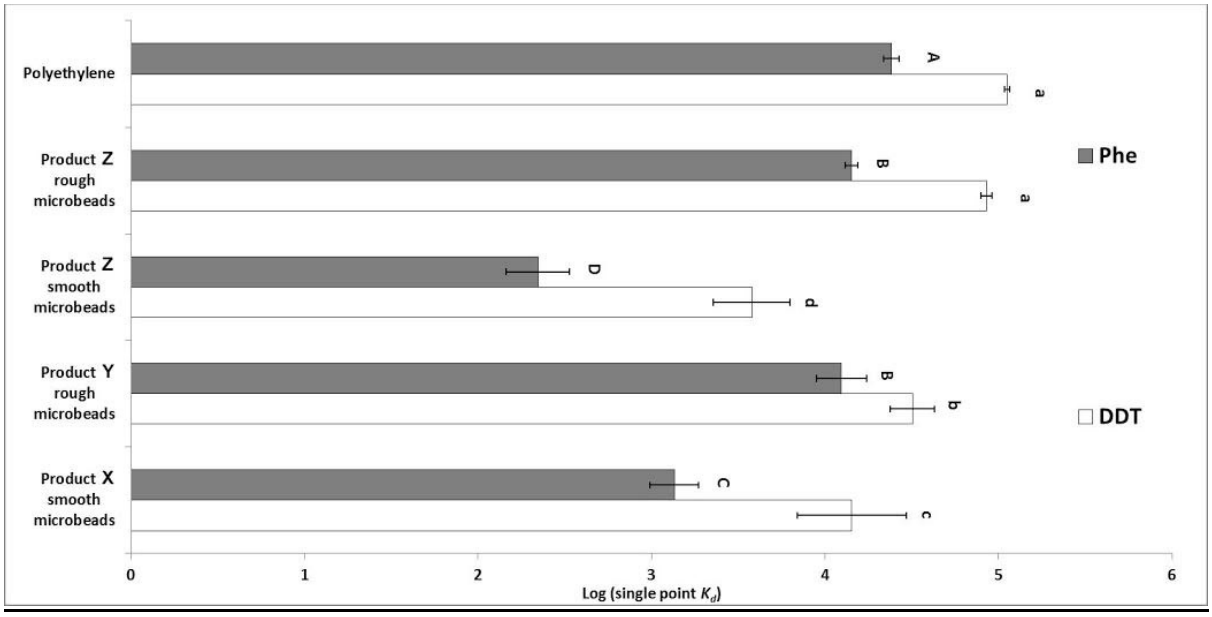
566



567

568 **Fig 4.** *A-Scanning electron microscopy (SEM) of a typical rough facial scrub plastic*
569 *microbead particle (9000X magnification). B- SEM of surface microbead*
570 *topography (16000X magnification). C- SEM of a broken smooth spherical*
571 *plastic microbead from 'product F' (900X magnification).*

572



573

574 **Fig 5.** *Single point distribution coefficients (K_d) for the sorption of a mixture of*
575 *phenanthrene (Phe) and DDT onto PE particles and rough and smooth PE-*
576 *microbeads extracted from cosmetic products (n=3, ± SD).* For each
577 contaminant, treatments with the same letters (A-C for Phe and a-d for DDT)
578 were not significantly different ($p < 0.05$).

579

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